Review

Ceramic and polymeric solid electrolytes for lithium-ion batteries

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ABSTRACT

Lithium-ion batteries are important for energy storage in a wide variety of applications including consumer electronics, transportation and large-scale energy production. The performance of lithium-ion batteries depends on the materials used. One critical component is the electrolyte, which is the focus of this paper. In particular, inorganic ceramic and organic polymer solid-electrolyte materials are reviewed. Solid electrolytes provide advantages in terms of simplicity of design and operational safety, but typically have conductivities that are lower than those of organic liquid electrolytes. This paper provides a comparison of the conductivities of solid-electrolyte materials being used or developed for use in lithium-ion batteries.

Contents

1. Introduction

Lithium-ion batteries are important for a wide variety of applications ranging from consumer electronics to automotive propulsion to stationary load-leveling for intermittent power generation, such as from wind or solar energy. Battery performance depends critically on the materials used, so the development of new materials is important for advancing battery technology [1,2]. One materials challenge is the development of electrode materials with increased energy density, faster discharge kinetics and better stability. Another materials challenge is the development of safer and more reliable electrolytes to replace the currently used organic carbonate liquid solutions. This latter challenge is the focus of this paper. In particular, the properties, especially conductivity, of solid electrolytes for lithium-ion batteries are reviewed.

The use of a solid electrolyte eliminates the need for containment of the liquid electrolyte, which simplifies the cell design, as well as improves safety and durability. There are two general classes of materials used for solid electrolytes in lithium-ion batteries: inorganic ceramics and organic polymers. The most obvious difference between these classes is the mechanical properties. The high elastic moduli of ceramics make them more suitable for rigid battery designs as in, for example, thin-film-based devices. Conversely, the low elastic moduli of polymers are useful for flexible battery designs. Polymers are also generally easier to process than ceramics, which reduces the fabrication costs. On the other hand, ceramics are more suitable for high temperature or other aggressive environments. One of the most important properties of electrolyte materials is the ionic conductivity and that is the focus of this paper. The paper contains a compilation of recent conductivity data from the literature, which are compared on Arrhenius-type plots. The purpose of this compilation is to determine the ranges of conductivity for various electrolyte materials and provide a basis for comparison between electrolyte systems.

2. Ceramic electrolytes

Ionic conduction in ceramic compounds occurs by movement of ionic point defects, the creation and/or movement of which requires energy, so the conductivity of these compounds increases with increasing temperature. Thus, ceramic solid electrolytes are well suited for high-temperature applications. However, ionic conduction in some compounds is reasonably high even at relatively low temperatures, so there are several types of lithium-ion conducting inorganic ceramics that have been investigated for use in lithium-ion batteries [3–7]. In this paper, the conductivity of sulfide, oxide and phosphate compounds for use as the electrolyte in lithium-ion batteries will be reviewed.

2.1. Sulfides

Sulfide compounds in crystalline, amorphous and partially crystalline forms have been used as lithium-ion conductors. One example is a $Li_2S–P_2S_5$ glass or glass–ceramic, the maximum conductivity for which occurs at 20–30% P_2S_5, depending on the degree of crystallization [8–10]. The conductivities of some $Li_2S–P_2S_5$ electrolytes are shown in Fig. 1 [10–12]. The conductivity of the glass–ceramics (i.e. after crystallization) are higher than those of the corresponding glasses at lower temperatures, which indicates that the crystalline phase has a higher conductivity and lower activation energy than the amorphous phase. This is not always the case, since in some other sulfide glasses (e.g. $Li_2S–P_2S_5–Li_4SiO_4$ [10]), crystallization decreases conductivity. Sulfide electrolytes have been used in batteries with lithium–indium anodes and several differ-

ent cathodes, including sulfides of nickel [13,14], copper [15] and copper–molybdenum [16], which operate at potentials of 2–3 V vs. Li/Li^+. Sulfide electrolytes have also been used in batteries with a NiP_2 cathode [17], which operates at a lower potential, as well as with more widely used cathodes, including $LiCoO_2$ [18–20] and $Li_4Ti_5O_{12}$ [21,22].

The conductivities of some other sulfide glasses are shown in Fig. 2 [10,11,23–25]. The conductivity of $Li_2S–SiS_2$ is similar to that of $Li_2S–P_2S_5$, but can be improved with lithium silicate (Li_4SiO_4) additions. The conductivity of $Li_2S–Ga_2S_3–GeS_2$ is similar to, or lower than, that of $Li_2S–P_2S_5$, but can be improved with LiI additions. $Li_2S–Sb_2S_3–GeS_2$, however, has a lower conductivity than $Li_2S–P_2S_5$.

Some lithium-ion-conducting sulfide crystals, which are referred to as thio-LISICON, have been reported and the conductivities of some are shown in Fig. 3 [26–30]. Crystalline $Li_{3.25}Ge_{0.25}P_{0.75}S_4$ has a high conductivity, which is in the same range as those of $Li_2S–P_2S_5$ glass–ceramics, but the conductivities of $Li_{4.2}Ge_{0.8}Ga_{0.2}S_4$ and $Li_{2.2}Zn_{0.1}Zr_{1.9}S_3$, are lower than those of $Li_2S–P_2S_5$ glass–ceramics. The higher conductivity of $Li_2S–GeS_2–P_2S_5$ has resulted in lower polarization and higher capacity with a $LiCoO_2$ cathode as compared to $Li_3PO_4–Li_2S–SiS_2$, but $Li_2S–GeS_2–P_2S_5$ was found to be incompatible with a graphite anode [31]. Like $Li_2S–P_2S_5$, $Li_2S–GeS_2–P_2S_5$ has been used in batteries with sulfide cathodes, such as Mo_6S_8, which operates at 2–3 V vs. Li/Li^+ [32]. The addition of oxygen to $Li_2S–GeS_2$ increases the room temperature conductivity [33]. The increase is attributed to an increase in the coordination number of lithium, but larger amounts of oxygen lead to a decrease in free volume, which results in a decrease in conductivity.

2.2. Oxides

Although GeO_2 has lower conductivity than GeS_2, the oxides in other systems are good lithium-ion conductors. One promising lithium-ion conducting oxide is the perovskite $(La,Li)TiO_3$ (LLTO) [34], the conductivities of which are shown in Fig. 4 [35–44]. Increasing the La/Li ratio above 1 results in the formation of A-site vacancies to maintain charge neutrality. The composition can be expressed as $Li_{3y}La_{2/3−y}TiO_3$, for which a maximum in conductivity has been observed at $x = 0.125$ (La/Li = 1.4) [45,46]. This is consistent with molecular dynamics calculations that indicate a maximum conductivity at $x = 0.105$ and minimum conductivity at $x = 0.045$ [47]. At lower lithium contents (small x) an orthorhombic phase forms, rather than the tetragonal phase, which forms at $x \sim 0.08$ and has a higher conductivity [48]. The conductivity results in Fig. 4 are separated into those for total conductivity and those for bulk grain conductivity. The higher bulk grain conductivity indicates that grain boundaries impede the transport of lithium ions. The results for total conductivity vary much more between different reports as compared to those for bulk conductivities, which may be due to variations in grain size and possibly grain boundary composition.

The effect of doping LLTO on the bulk conductivity is shown in Fig. 5 [43,44,49,50]. Small amounts of aluminum replacing titanium have been shown to lead to high conductivity, but the addition of sodium, which is added to the A-site (i.e. replaces Li^+/La^{3+}), leads to a decrease in conductivity. The replacement of some oxygen with fluorine does not significantly affect the conductivity. Other studies have shown that the addition of silica to $Li_{0.5}La_{0.5}TiO_3$ increases total conductivity, which could be due to its effect on grain boundary transport [35]. LLTO has also been mixed with $LiMn_2O_4$ to form a composite cathode [51].

Some oxides forming a garnet-related structure also have good lithium-ion conductivity. The conductivities of some garnet oxides based on $Li_5La_3Ta_2O_{12}$ are shown in Fig. 6 [52–57]. The conduc-
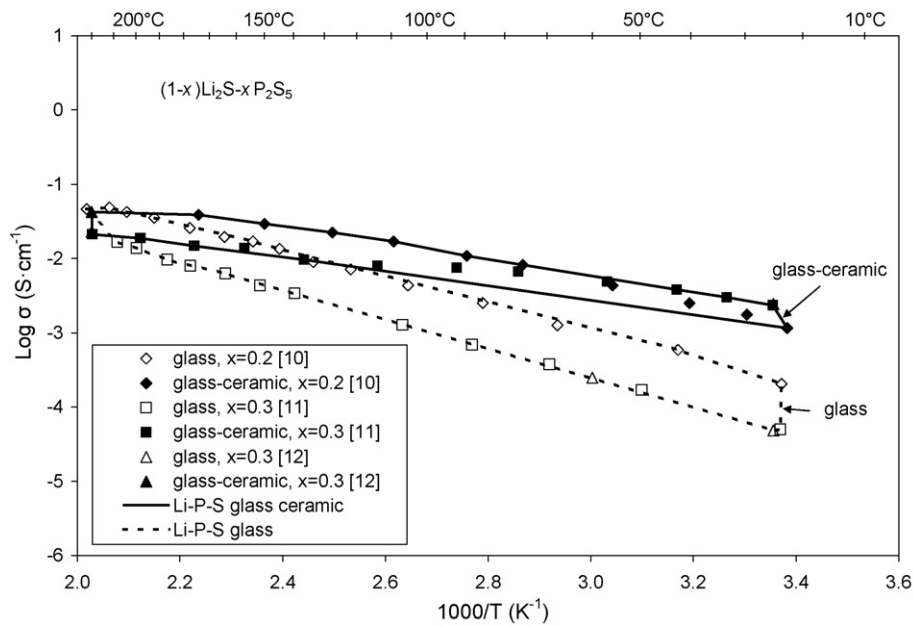
Fig. 1. Conductivity of Li_2S–P_2S_5 [10–12].

tivities of these garnet-based oxides are similar to those of LLTO. Replacing lanthanum with barium and/or strontium increases the conductivity. Calcium, however, does not have the same beneficial effect on conductivity, since, as shown in Fig. 7 [56,58–60], the conductivity of $Li_6La_2CaTa_2O_{12}$ is lower than those of the barium- or strontium-doped garnets. Fig. 7 also shows that the niobates, *e.g.* $Li_6La_2ANb_2O_{12}$ (A = Ca, Sr), have lower conductivities than the tantalates and that strontium is more beneficial than calcium as a dopant in both systems [58]. The conductivity of the garnet, $Li_2Nd_3TeSbO_{12}$, has also been reported, but is lower than those shown in Fig. 7 [59,60]. Fig. 7 also shows that the conductivities of garnet oxides based on niobium or antimony are not as high as those of barium- or strontium-doped lanthanum tantalates.

The conductivities of several other oxides reported as potential lithium-ion solid electrolytes are shown in Fig. 8 [61–66]. The conductivities of a $Li_3BO_{2.5}N_{0.5}$ thin-film glass and of Li_9SiAlO_8, which is a derivative of Li_4SiO_4, are close to the conductivity range of LLTO. Other oxides, however, have conductivities that are considerably lower than LLTO. These low conductivities may preclude the use of these materials as the electrolyte in a conventional room temperature battery, but materials with low conductivities may still be used in thin-film or high-temperature batteries. For example, although the conductivity of $Li_3BO_{2.5}N_{0.5}$ is lower than that of LLTO at room temperature it has been used in a thin-film battery [64]. Some oxides that have low conductivity room temperature conductivity, but have potential for use at higher temperatures include silicates [67–70] and
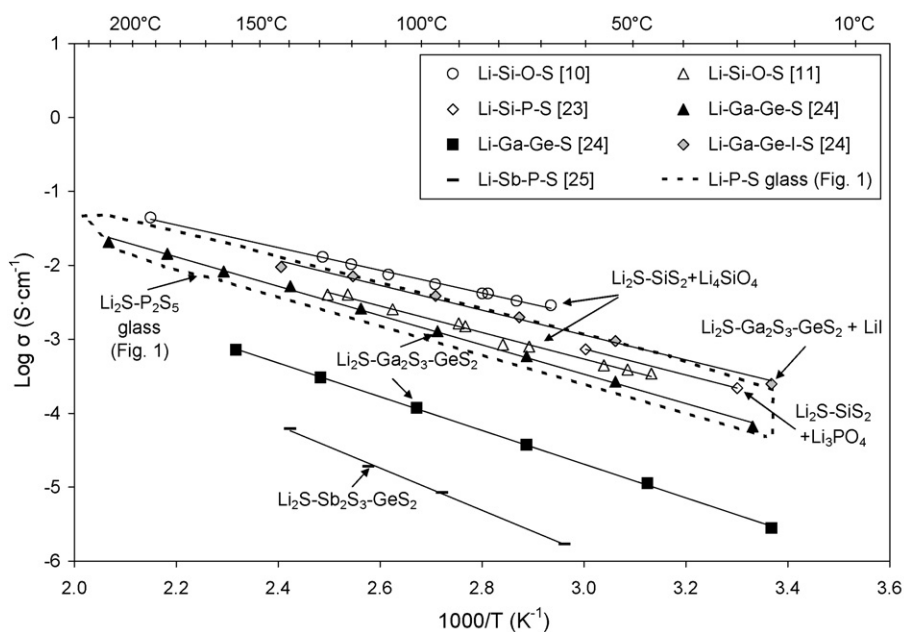
Fig. 2. Conductivity of other sulfide glasses [10,11,23–25].
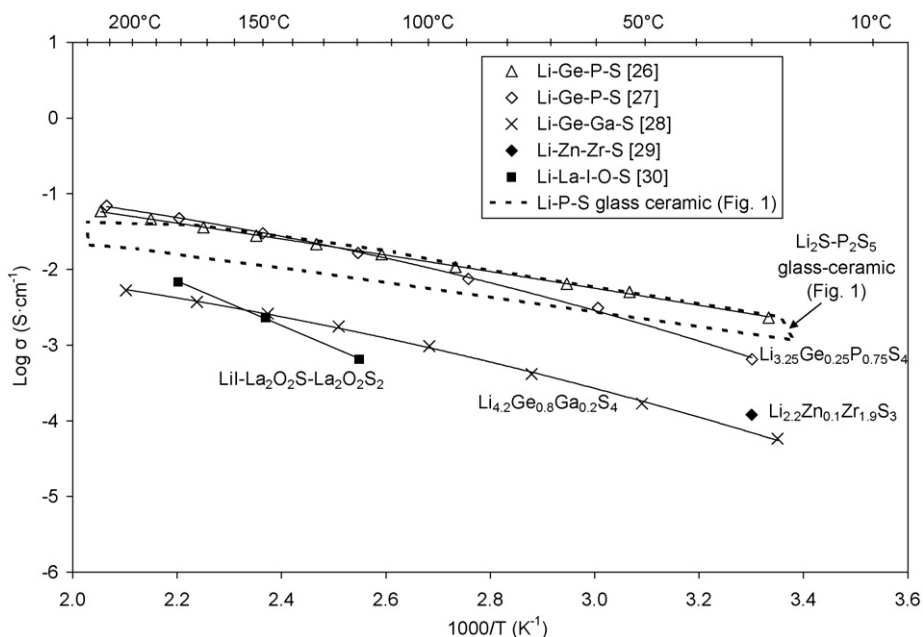
Fig. 3. Conductivity of other sulfide crystals [26–30].

titanates, such as $Li_{2-2x}Mg_{2x}TiO_{3+x}$ [71] and $Li_{2x}Zn_{2-3x}Ti_{1+x}O_4$ [72].

2.3. Phosphates

Some phosphates, similar to the sodium-ion conducting NASICON compounds, are good lithium-ion conductors. The conductivities of some of these lithium-ion-conducting phosphate compounds are shown in Fig. 9 [73–85]. The highest conductivities are observed for $Li_{1+x}Al_xGe_{2-x}(PO_4)_3$ (LAGP). Another commonly used phosphate electrolyte is $Li_{1+x}Ti_{2-x}Al_x(PO_4)_3$ (LTAP), to which silicon has been added to replace some of the phosphorous (*i.e.* $Li_{1+x+y}Ti_{2-x}Al_xSi_y(PO_4)_{3-y}$). LTAP has been used as the electrolyte in lithium-ion batteries [86,87]. The conductivity of the zirconium-based phosphate, $LiTi_{0.5}Zr_{1.5}(PO_4)_3$, is lower than those of the titanium- and germanium-based phosphates.

The conductivities of some other phosphate electrolytes are shown in Fig. 10 [84,89–96]. As shown above for LLTO (Fig. 4), the bulk conductivity, at least in some cases, is higher than the total conductivity. The bulk conductivity of the zirconia-based phosphate, $LiTi_{0.5}Zr_{1.5}(PO_4)_3$, is lower than that of LTAP, which is consistent with impedance spectroscopy studies of $LiTi_{2-x}Zr_x(PO_4)_3$ indicating that zirconium additions affect the bulk conductivity more than the grain boundary conductivity [97]. However, zirconia-based phosphates, *e.g.* $Li_xAlZr[PO_4]_3$ are being investigated for higher temperature applications [98]. Fig. 10 also shows that lithium phosphate glass has a relatively low conductivity, but its conductivity can be increased with the addition of aluminum or halogen salts. Glasses with $Li_3Fe_2(PO_4)_3$ crystals have been mentioned as potential electrolyte materials [99], but transition metal phosphates are more commonly used for cathode materials in lithium-ion batteries, because the change in valence
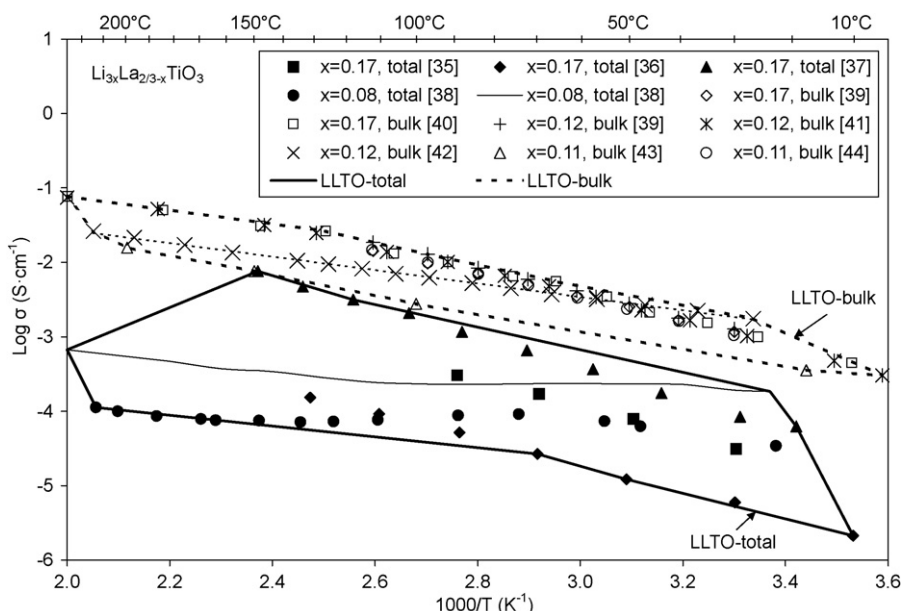
Fig. 4. Conductivity of $Li_{0.5}La_{0.5}TiO_3$ (LLTO) [35–44].
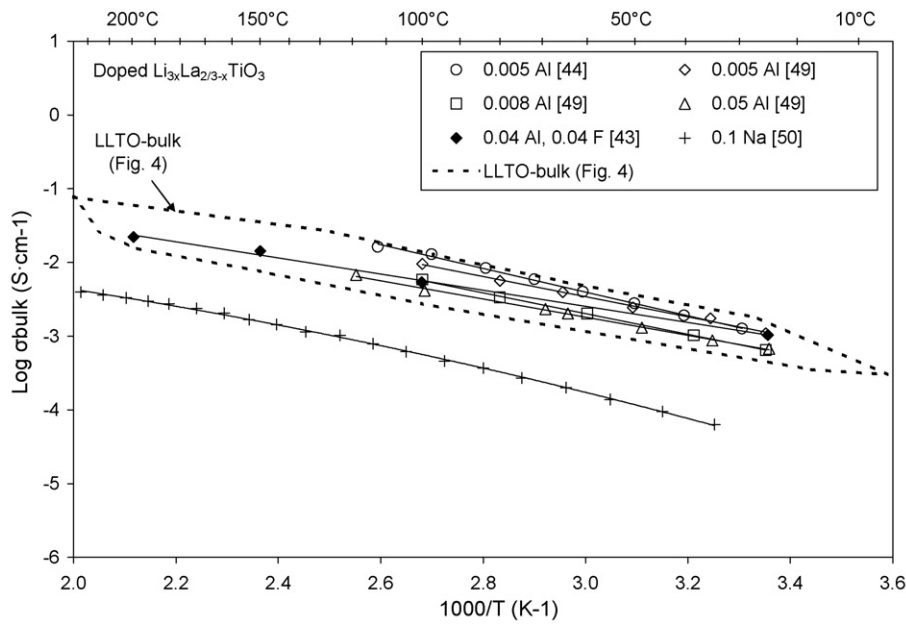
Fig. 5. Bulk grain conductivity of LLTO with Al, Na and/or F additions [43,44,49,50].

of the transition metal allows for insertion and removal of lithium ions [100]. Another phosphate-related compound is LiPON, which is a lithium-ion conductor, but has a low conductivity (*i.e.* $\sim 10^{-5}$ to $10^{-6}\,\mathrm{S\,cm^{-1}}$) [101]. However, reducing the electrolyte thickness reduces the resistance, so LiPON has been used in thin-film batteries [102–109].

3. Polymer electrolytes

Polymer electrolytes offer advantages over ceramics, including good processibility and flexibility, while maintaining the advantages of solid electrolytes, including dimensional stability, safety and the ability to prevent lithium dendrite formation [110–113]. In some polymer electrolytes, lithium salts are solvated by the polymer chains, while in others a solvent is added to form a polymer

gel. In the general, the former is mechanically stronger, so that a free-standing film can be formed. Polymer gels, on the other hand, require mechanical support from other battery components, but typically have higher conductivities. Examples of both types will be discussed below.

3.1. Solid polymer electrolytes

The polymer most commonly used for lithium-ion conducting electrolytes is poly(ethylene oxide) (PEO). PEO is effective in solvating lithium salts, which are added to provide the lithium-ion conduction. The conductivities of PEO with different salts are show in Figs. 11 and 12, where results for lithium trifluoromethanesulfonate ($LiCF_3SO_3$) (LiTf) lithium bis(trifluoromethanesulfonimidate) ($Li(CF_3SO_2)_2N$) (LiTFSI) and
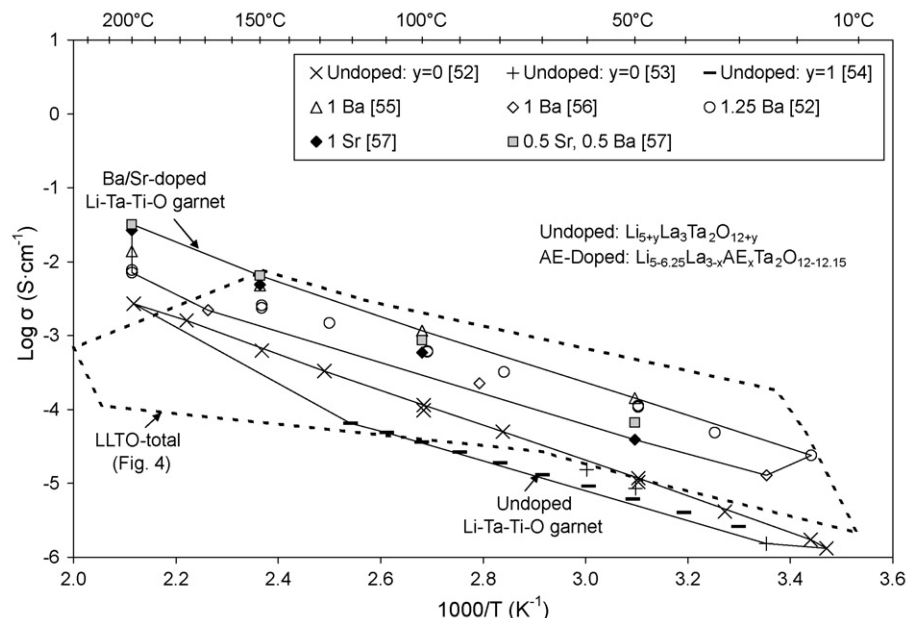
Fig. 6. Conductivity of tantalum-based garnet oxides [52–57].
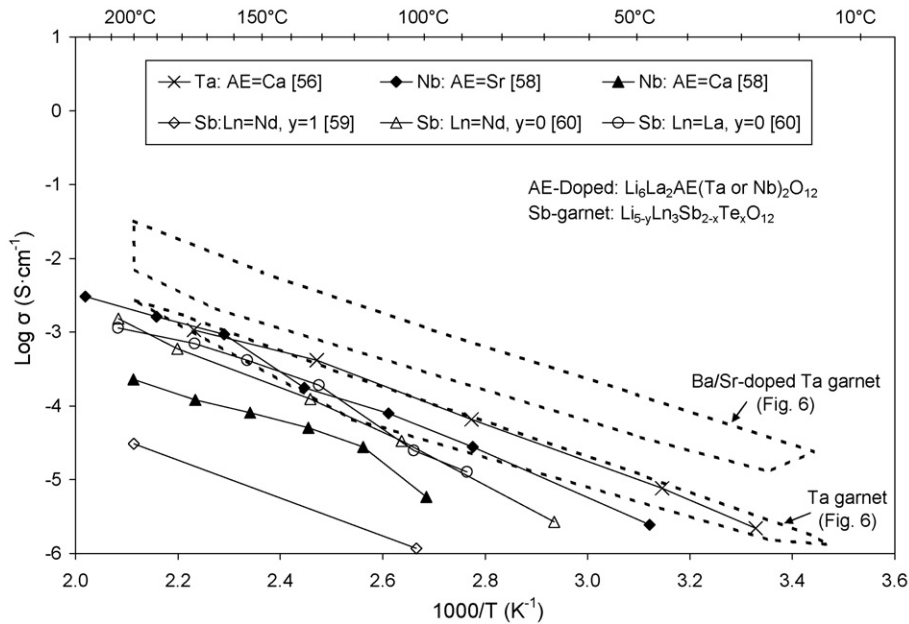
Fig. 7. Conductivity of selected garnet oxides [56,58–60].

lithium bis(trifluoromethanesulfonimide) $(Li(C_2F_5SO_2)_2N)$ (LiBETI) are shown in Fig. 11 [82,114–122], and results for lithium perchlorate $(LiClO_4)$ and lithium bis(oxalato)borate $(LiB(C_2O_4)_2)$ (LiBOB) are shown in Fig. 12 [123–127]. The conductivities of PEO with the different salts are in a similar range, with the highest conductivities occurring for LiTFSI and the lowest for LiBETI. There is one report [121] of a conductivity that is orders of magnitude higher than all other reported values. In that paper, the activation energy was compared with other reports from the literature, but the magnitude was not. Since the reason for this high conductivity is not clear, this result was not included in the representation of the range of conductivity values for PEO. The decrease in activation energy for lithium-ion conduction in PEO with increasing temperature is associated with heating above the glass transition temperature.

Although PEO is a good lithium-ion conductor, higher conductivities are needed for room temperature battery applications.

The good ionic conductivity of PEO is due to transport in the amorphous region, so the conductivity decreases with increasing degree of crystallization. Thus, one approach to increasing the conductivity is to add a plasticizer, such as succinonitrile (SN) [122], polysquarate (PPS) [117] or ethylene carbonate (EC)/propylene carbonate (PC) [114], which reduces crystallization and thus increases conductivity, but the conduction mechanism does not change [128]. The conductivities of PEO with plasticizers, as well as some other modifications, are shown in Fig. 13 [114,117,118,122,124,126,129–133]. PEO chains can be incorporated in other polymers, such as urethane/siloxane [133] to combine the solvating properties of PEO in an amorphous matrix.
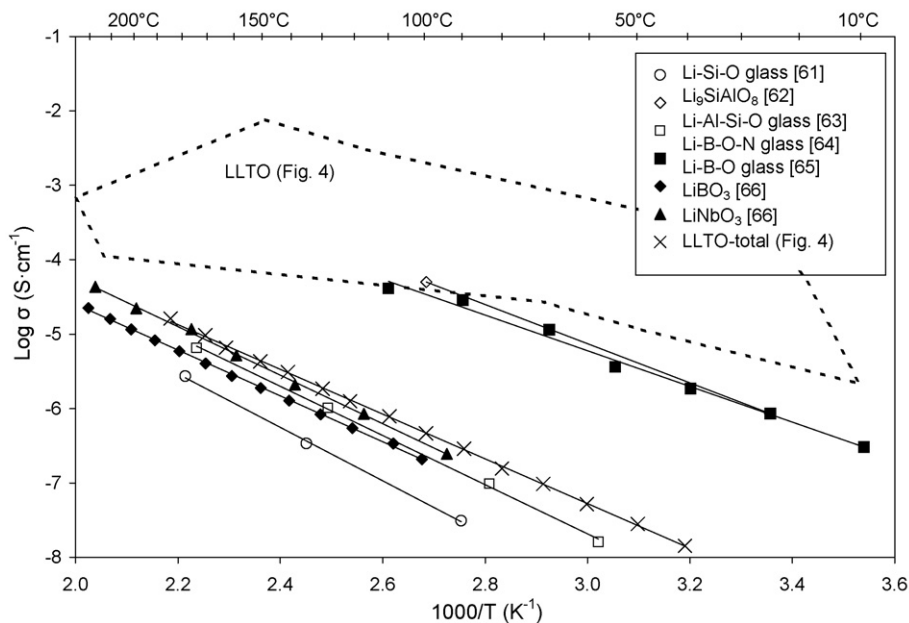
Fig. 8. Conductivity of other oxides [61–66].
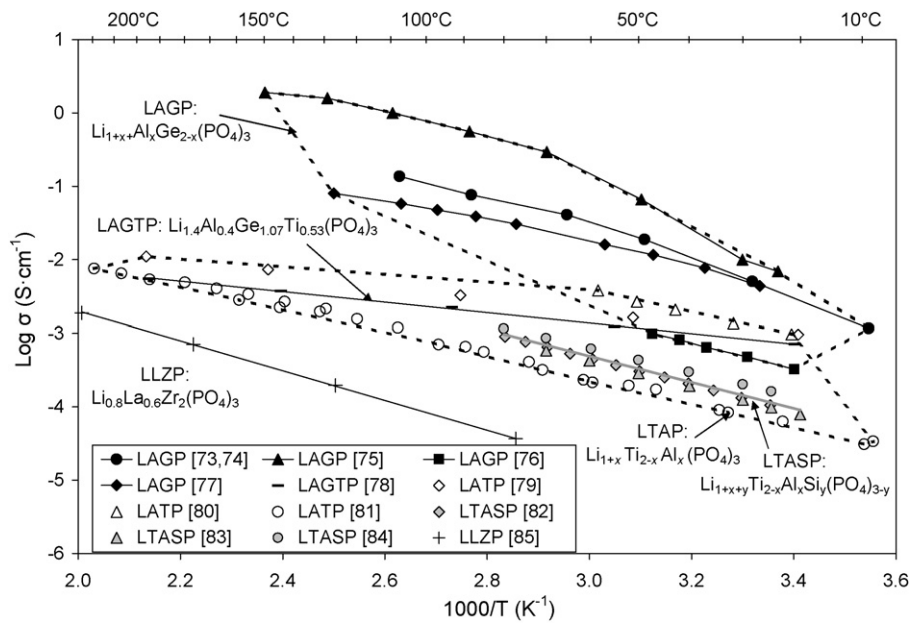
Fig. 9. Conductivity of phosphates [73–85].

Another approach is to add a room temperature ionic liquid (RTIL), such as 1-ethyl-3-methylimidazolium (EMI) [118] or N-methyl-N-butylpyrrolidinium bis (trifluoromethansulfonyl) imide (PYTRA$_{14}$TFSI) [130], to weaken the interaction between the lithium ions and the polymer chains to increase lithium-ion mobility. The interaction with lithium ions can also be affected by forming a copolymer. For example, the addition of polymers with carboxylic acid group, such as poly(acrylic acid) (PAA) and poly(methacrylic acid) (PMAA), leads to an increase in the conductivity of PEO [124]. The addition of PAA or other poly(lithium carboxylate)s can also increase the lithium-ion transport number by reducing the transport of anions [134]. The conductivity of such polymers can be improved with the addition of boron trifluoride etherate (BF$_3$·OEt$_2$), which promotes dissociation of the lithium cations and carboxylate anions. Epoxidized natural rubber (ENR) additions have also been

shown to increase the conductivity of PEO [132,135]. The addition of poly(styrene-block-ethylene oxide) (SEO) to PEO has been shown to affect the distribution of lithium ions, such that the conductivity increases with increasing molecular weight, which is the opposite of the typical behavior of polymer electrolytes [136].

The addition of ceramic particles, such as alumina [114], silica [126,137] and CuO [129], leads to an increase in conductivity. These ceramic particles can lead to decreased crystallization, but may also enhance conduction at the polymer-ceramic interface. For example, improvements using a γ-glycidoxypropyltrimethoxy silane (GPTMS) treatment of mesoporous silica (SBA) particles have been attributed to reduced crystallization and to the interaction between the modified surface and PEO polymer chains [126]. The addition of ceramic particles can also improve the strength of the polymer, which is important in designing a polymer electrolyte,
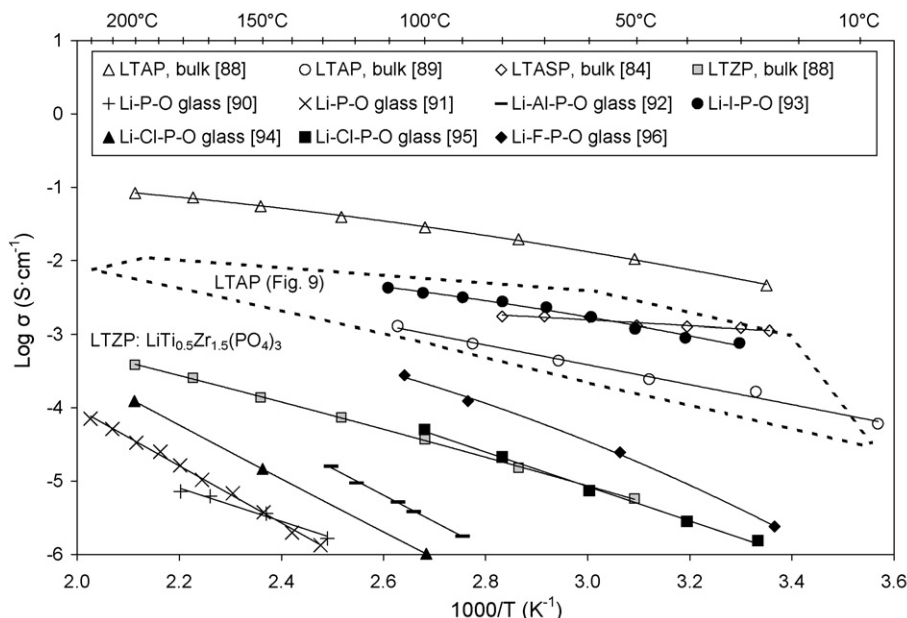
Fig. 10. Conductivity of phosphates [84,88–96].
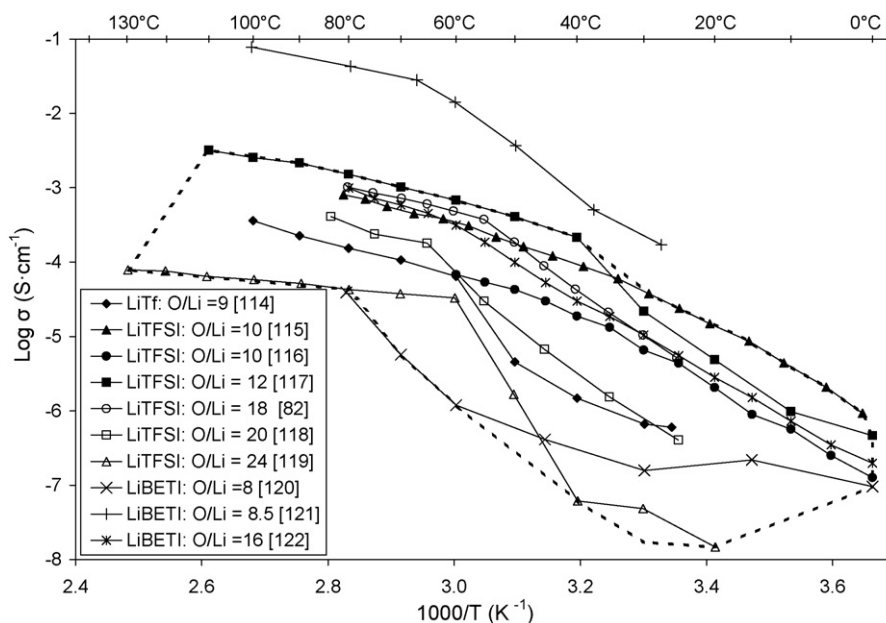
Fig. 11. Conductivity of PEO with LiTf, LiTFSI or LIBETI salts [82,114–122].

because most changes that increase conductivity are detrimental to mechanical performance [138]. PEO is stable with lithium metal [139] and has been demonstrated in a battery to have a capacity that is similar (~95%) to a battery with the commonly used EC/DEC/LIPF$_6$ electrolyte [140]. PEO electrolytes have been used in batteries with various electrodes including LiMn$_2$O$_4$ [141] and LiFePO$_4$ [142–144] cathodes, as well as carbon [145] and MoO$_x$ [146,147] anodes.

In addition to modifying PEO, alternative solid polymer electrolyte materials have been developed. Fig. 14 [148–157] shows that the conductivities of other solid polymer electrolytes are typically in the same range as those of PEO, which is indicated by the broken lines. Electrolytes containing ethylene oxide, such as poly(ethylene oxide) methyl ether methacrylate (PEOMA) [148] and poly(acetyl-oligo(ethylene oxide) acrylate) (PAEOA) [149], or poly(ethylene glycol) (PEG), such as polyester diacrylate

(PEDA)-PEG copolymers [150], poly(ethylene glycol) dimethacrylate (PDE) [152], poly(ethylene glycol) methacrylate (PME) [152], poly(ethylene glycol) methyl ether methacrylate (PEGMA) [153] and tri(ethylene glycol) diacrylate (TEGDA) [152], have conductivities in the range of, or even higher than, PEO.

The design of polymer electrolytes often uses complementary properties of the constituents. For example, the conduction properties of PME (which contains an ethylene oxide chain) can be combined with the strength of the self-assembled copolymer, stearylmethacrylate (SMA) [151]. The two components can also combine conduction mechanisms as in the combination of the segmental polymer chain motion mechanism of PEOMA with the lithium-ion hopping mechanism of α-methyalene γ-butyrolactone (MBL) [148]. The different conduction mechanisms can dominate in different temperature ranges, which expands the useful operating
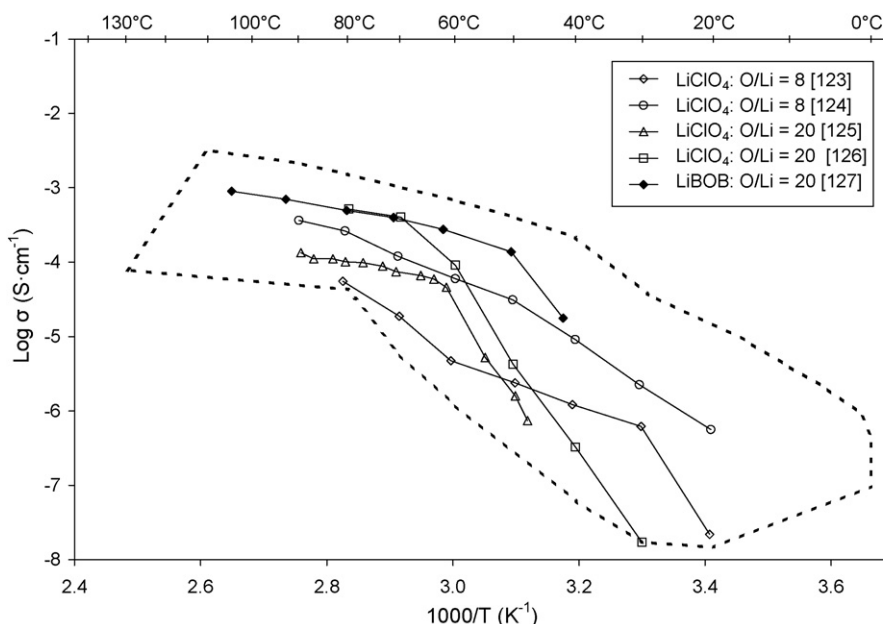
Fig. 12. Conductivity of PEO with LiClO$_4$ or LiBOB salts [123–127].
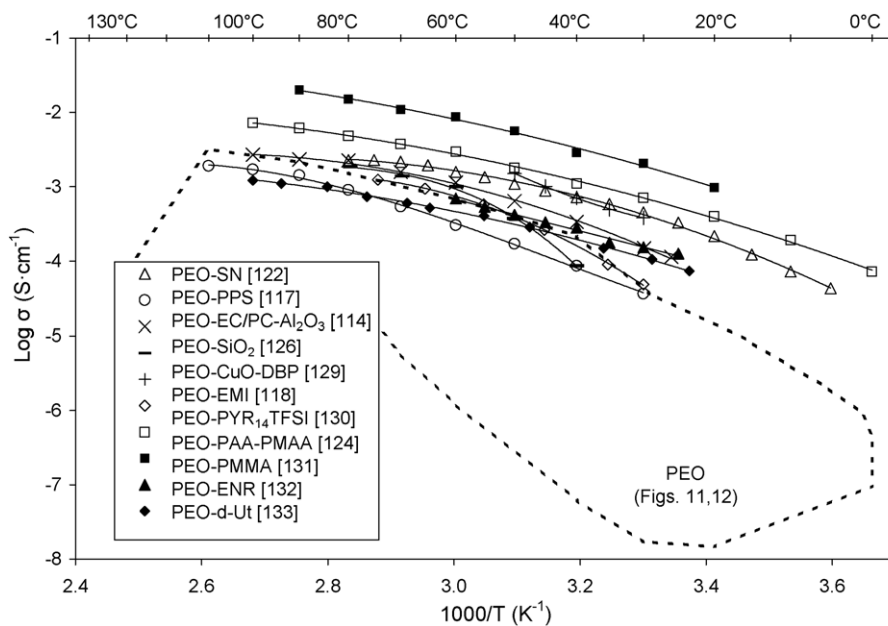
Fig. 13. Conductivity of PEO with additives [114,117,118,122,124,126,129–133].

temperature of the electrolyte. For example, in the case of PEOMA-MBL, the dominant mechanism shifts from segmental polymer chain motion to lithium-ion hopping with increasing temperature. Lithium-ion transport can also be enhanced by forming a lamellar structure, such as in a polystyrene (PS)-poly(2-vinylpyridine) (P2VP) block copolymer [155].

3.2. Polymer gel electrolytes

Another approach for polymer electrolytes is to form a polymer gel by incorporating an organic liquid electrolyte in the polymer matrix [158]. The conduction mechanism in polymer gels is similar to that in the liquid electrolytes, but a gel has improved safety and shape flexibility over a liquid. The most commonly used polymer gel electrolyte is poly(vinylidene fluoride) (PVdF) to which hexaflu-oropropylene (HFP) is typically added to reduce the crystallinity of the polymer. The conductivities of PVdF-HFP gel electrolytes with $LiPF_6$ as the lithium salt and carbonate solvents, EC, PC, dimethyl carbonate (DMC) and ethyl methyl carbonate (EMC), are shown in Fig. 15 [159–167]. Not included in Fig. 15 is a recent report of a conductivity of more than $4\,S\,cm^{-1}$, which is more than two orders of magnitude larger than any other reported values [168]. As in the case discussed above for PEO shown in Fig. 11, in that paper, the activation energy, but not the absolute value of conductivity, was compared with results from the literature, so the reason for this high value of conductivity is not clear. In general, the conductivities of PVdF-HFP with EC-PC as the solvent are higher than those with EC-DMC as the solvent.

The conductivity of gel electrolytes can be increased by improving the pore structure to facilitate transport of lithium in the
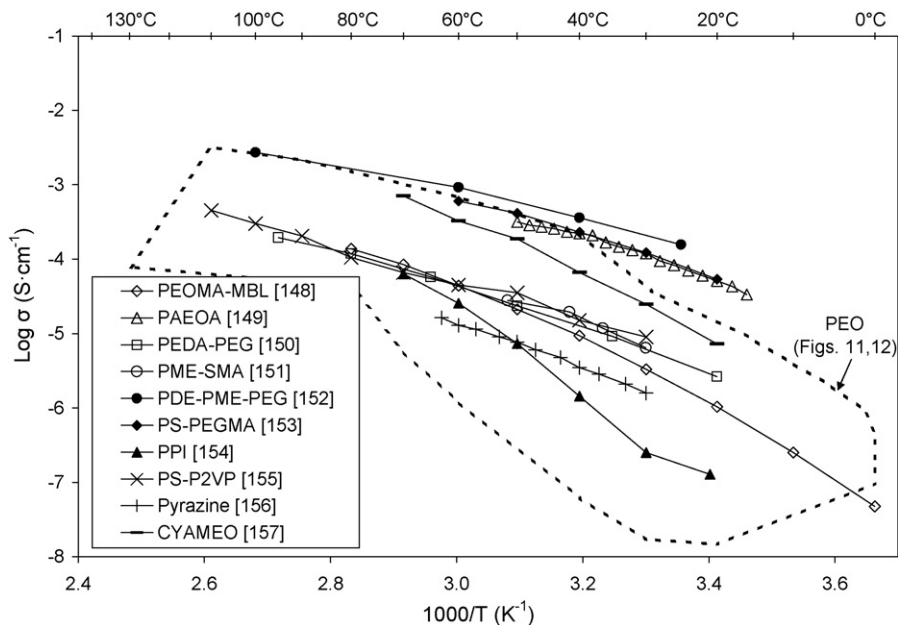
Fig. 14. Conductivity of other solid polymer electrolytes [148–157].
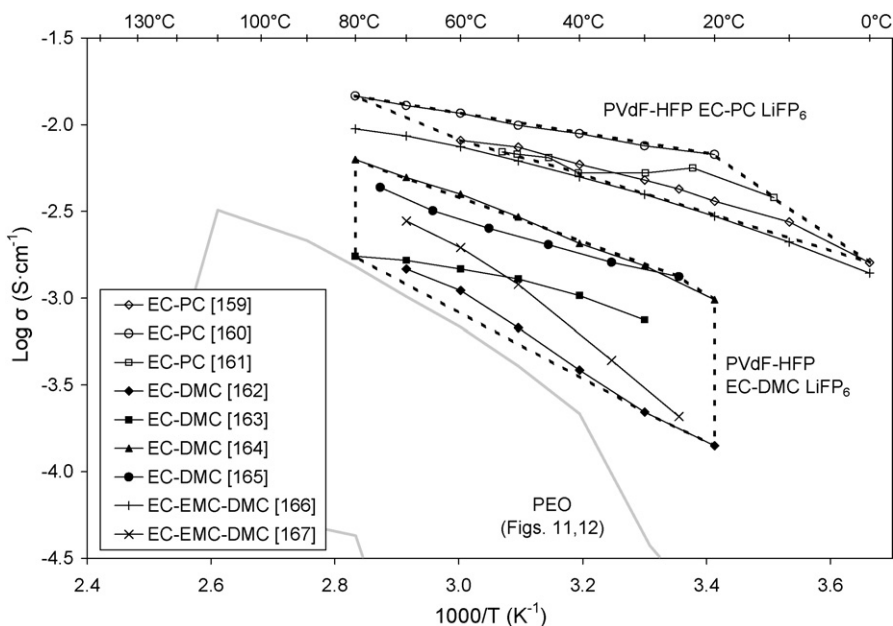
Fig. 15. Conductivity of PVdF-HFP with carbonate solvents [159–167].

solvent. This can be accomplished by adding ceramic particles, such as $BaTiO_3$ [159] or TiO_2 [162,163], or by modifying the polymer structure, such as with the addition of crosslinked dipoxy polyethylene glycol (DIEPEG) [164]. The conductivity of PVdF-HFP electrolytes with these and other additives are shown in Fig. 16 [159,162–164,167].

Another approach for electrolytes in lithium-ion batteries is the use of room temperature ionic liquids (RTIL), which can be used to replace the traditional organic solvent-based electrolytes. Ionic liquids are neither volatile nor flammable, which improves the safety of the battery during use. In addition, RTILs can be incorporated in polymers [169,170] or ceramics [171] to form a solid electrolyte. Fig. 17 [172–174] shows the conductivities of some PVdF-HFP electrolytes to which ionic liquids, N-n-butyl-N-ethylpyrrolidinium N,N-bis(trifluoromethane)sulfonimide ($Py_{24}TFSI$) [172], N-butyl-

N-ethylpiperidinium N,N-bis(trifluoromethane sulfonimide) ($PP_{24}TFSI$) [173] or N-methoxyethyl-N-methylpyrrolidinium bis(trifluoromethanesulfonyl)-imide ($PYRA_{12O1}TFSI$) [174], were added. The conductivities of the RTIL-based PVdF-HFP electrolytes are similar to those with $LiFP_6$ and a carbonate solvent. Polymerized ionic liquids have also been used as electrolytes or hosts for other ionic liquids, although the conductivities of these electrolytes are typically lower than when the ionic liquids are incorporated in a PVdF-HFP matrix [175,176].

One of the issues with the application of ionic liquids as electrolytes in lithium-ion batteries is the transport (or transference) number, which is the proportion of the current carried by a particular species. Any charge carried through the electrolyte by species other than lithium does not pass through the external circuit and thus does not provide electrical energy. In a ceramic electrolyte,
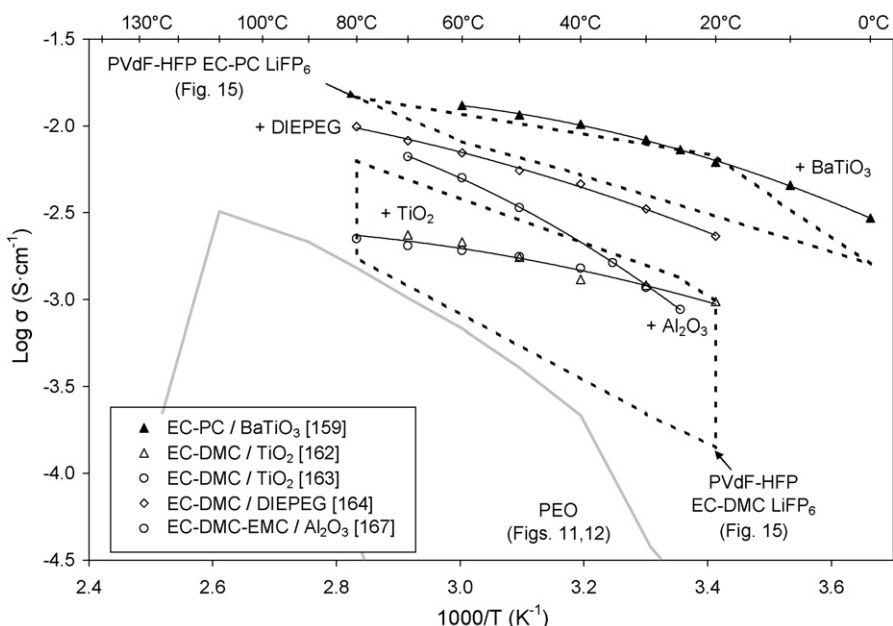
Fig. 16. Conductivity of PVdF-HFP with carbonate solvents and additives [159,162–164,167].
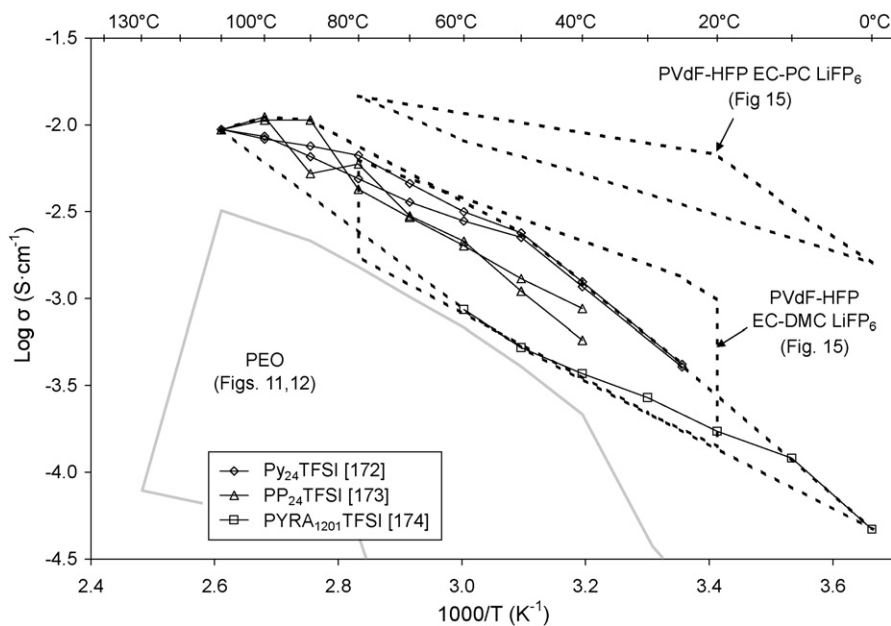
Fig. 17. Conductivity of PVdF-HFP with ionic liquid LiTFSI [172–174].

there is typically only one mobile ion, although conduction by electronic charge carriers (*i.e.* electrons and hole) can occur in some materials and conditions. In an ionic liquid, however, ions other than lithium can be mobile and thus carry charge [169,177]. In some cases, the conduction from lithium ions can be only a few percent of the total conduction [178]. The lithium-ion transport number of polymer electrolytes with ionic liquids can be similarly low. For example, lithium-ion transport numbers of 0.034 [179] and 0.06 [174] have been reported for PVdF-based electrolytes containing ionic liquids. However, the addition of plasticizer, such as EC [179], or ceramic particles, such as silica [174], can increase the transport number by several times.

The conductivities of PVdF-HFP electrolytes with other salts, LiBOB [180], LiTFSI [181] and LiClO$_4$ [182,183] are shown in Fig. 18 [172,180–183], and are in the same range as those with LiPF$_6$ as the salt. As with PEO-based electrolytes, the addition of ceramic particles can increase the conductivity, but large amounts of particles can degrade the mechanical properties [158]. The PVdF-HFP-PVC electrolyte in Fig. 18, which contains 17% PVC is reasonably high, but other reports for electrolytes with higher PVC contents (*e.g.* 30%) indicate a much lower conductivity (less than $10^{-4.5}$ S cm^{-1}) [184,185]. Additives can also improve the performance during battery operation. For example both organic (*e.g.* thiophene, 3,4 ethylenedioxythiophene (EDOT), biphenyl) [186] and inorganic additives (AlF$_3$, LiF) [187] improve the stability during battery cycling. Additives such as silica fibers can also help to improve the electrode capacity [188]. PVdF has been used with the common lithium-ion battery cathode materials, including LiCoO$_2$ [186,189], LiMn$_2$O$_4$ [190,191], LiFePO$_4$ [188] and LiNi$_{0.8}$Co$_{0.2}$O$_2$ [192], as well as in lithium-air batteries [193].
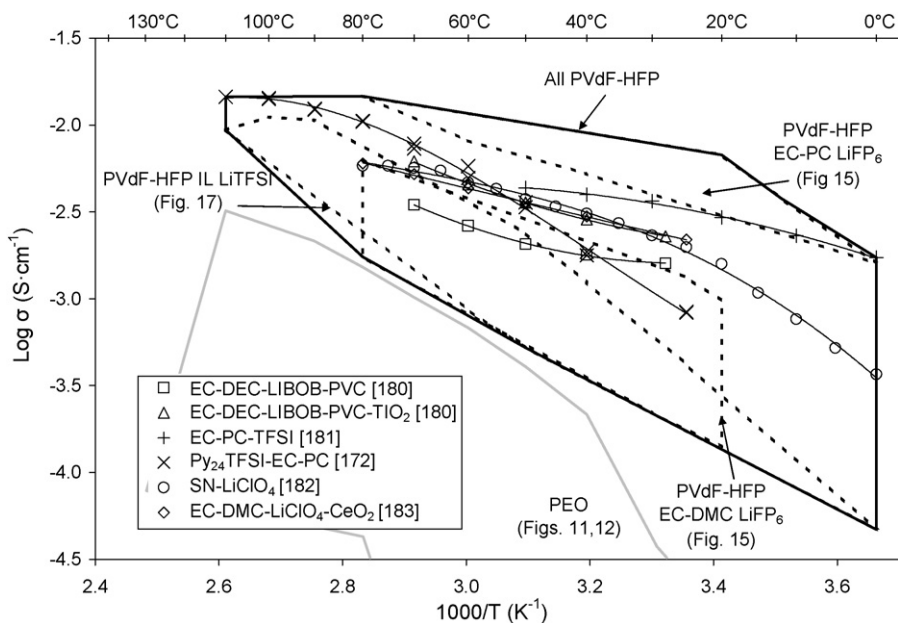
Fig. 18. Conductivity of PVdF-HFP with other additions [172,180–183].
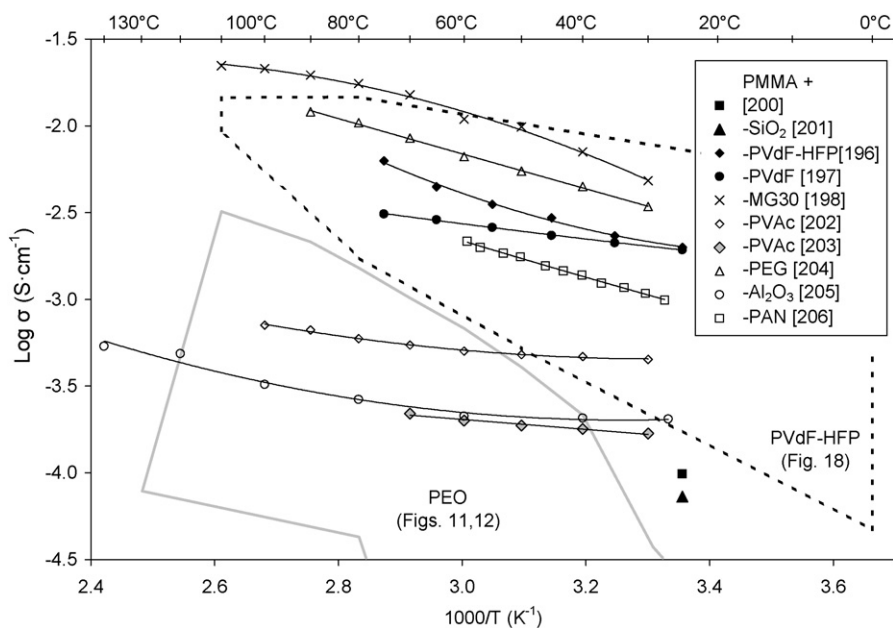
Fig. 19. Conductivity of PMMA-based electrolytes [196–198,200–206].

Another polymer used in gel electrolytes is poly(methyl methacrylate) (PMMA), which has a low conductivity (especially below 80 °C) [194] (*e.g.* 2.3×10^{-7} S cm^{-1} at room temperature) [195]. However, a layer of PMMA can be combined with PVdF to reduce leakage of the electrolyte solution [196,197]. In addition, PMMA is used in methyl-grafted natural rubber (MG30) to produce an electrolyte with high conductivity [198,199]. The conductivities of PMMA with MG30 and other PMMA-containing polymers are shown in Fig. 19 [196–198,200–206]. The conductivity of PMMA alone [200] or with poly(vinyl acetate) (PVAc) are relatively low [202,203] as is the conductivity when PMMA is used with PEO in an ionic liquid gel [207]. However, the conductivity of PMMA with poly(ethylene glycol) (PEG) is relatively good [204]. A copolymer of poly(acrylonitrile) (PAN) and PMMA has conductivity similar to

those of PVdF-HFP, and has been demonstrated in a battery with a lithium anode and a LiCoO$_2$ cathode to perform similarly to a battery with a liquid electrolyte [208].

PAN is a commonly used polymer for lithium-ion conducting electrolytes and the conductivities of some PAN-based electrolytes are shown in Fig. 20 [206,209–214]. The PAN-based electrolytes with the best conductivities are those with a LiClO$_4$ salt and SN, the conductivity of which is included for comparison. Succinonitrile is an organic plastic, which is an effective addition even at temperatures where its conductivity is low (*i.e.* below its melting point). As with PMMA, mixing PAN with PVC leads to a decrease in conductivity. The lithium-ion conductivity of PVC with additions of dibutyl phthalate (DBP) [215], ENR [216,217], PEG [218,219] or poly(oxyethylene)$_9$ methacrylate (POEM) [220] are in the range
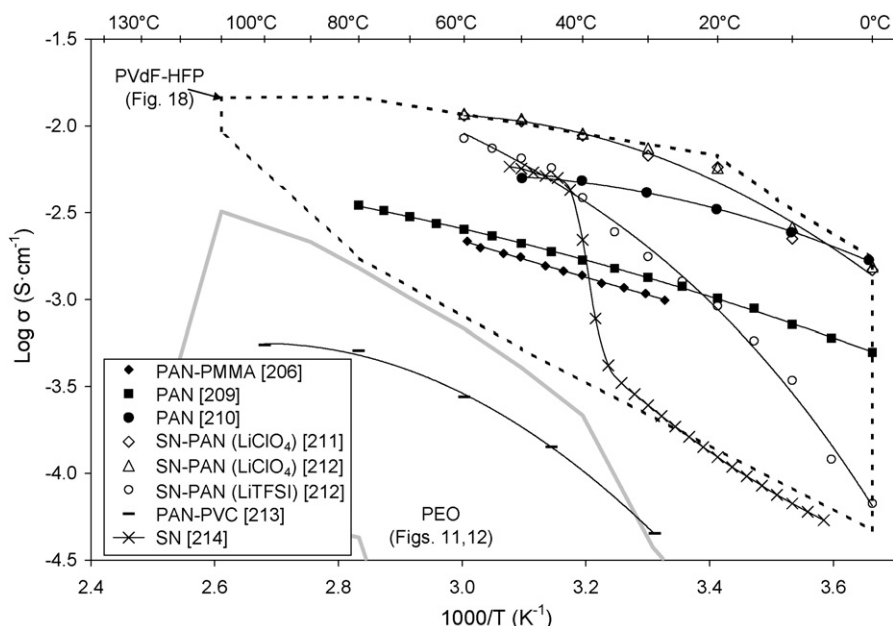
Fig. 20. Conductivity of PAN-based electrolytes [206,209–214].
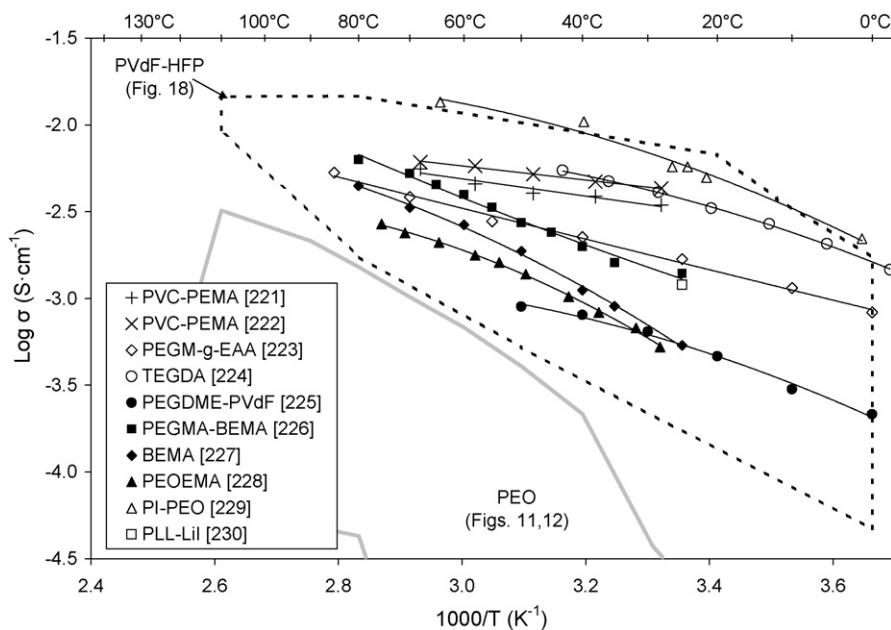
Fig. 21. Conductivity of other gel electrolytes [221–230].
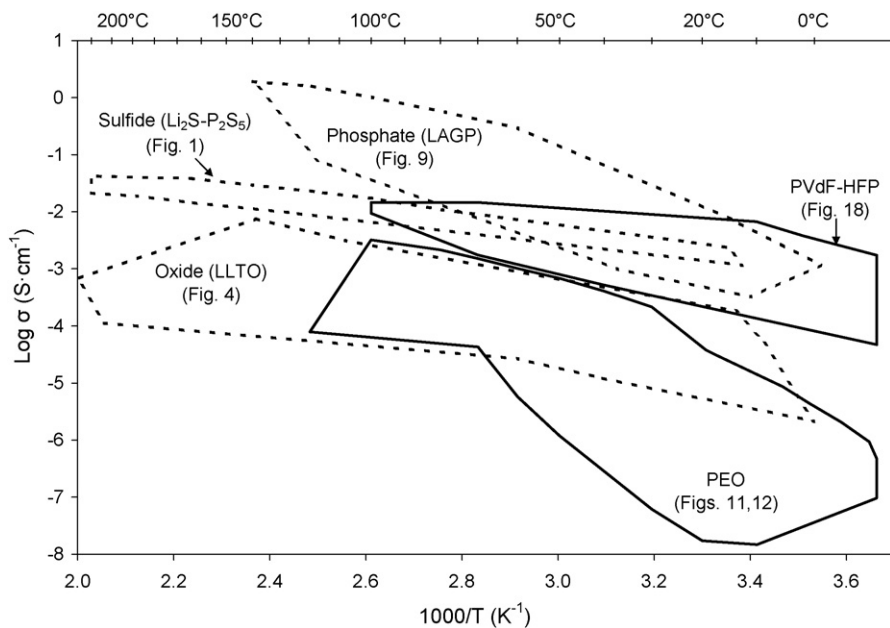
Fig. 22. Conductivity of inorganic ceramic and organic polymer lithium-ion conducting electrolytes.

or below those of PEO electrolytes (rather than PVdF-HFP gel electrolytes). However, the conductivity of PVC with poly(ethyl methacrylate) (PEMA) is comparable to PVdF-HFP as shown in Fig. 21 [221–230].

Ethylene glycol-based electrolyte, such as TEGMA, PEGMA and PEGM-g-EAA, which is a graft copolymer of polyethylene-poly(acrylic acid) copolymer esterified with poly(ethylene glycol)monomethyl ethers, have been used as lithium-ion conducting electrolytes and have shown conductivities that are in the range of those of PVdF-HFP. Dimethacrylic oligomers, such as bisphenol A ethoxylate (15 EO/phenol) dimethacrylate (BEMA), have been used alone [227] or as additions to PEGMA [226] as lithium-ion conducting electrolytes. Poly(2 ethoxyethyl methacrylate) (PEOEMA) is in the lower range of conductivity in Fig. 21 (although still in the range of PVdF-HPF), but has shown stability at potentials as

high as 5.1 V relative to Li/Li+ [228]. The highest conductivity in Fig. 21 is for a copolymer of polyimide (PI) and PEO with N-propyl-N-methylpyrrolidinium trifluoromethanesulfonimide (P_{13}TFSI) as a RTIL [229]. The high conductivity was achieved by increasing the amount of liquid (RTIL and EC/PC) to four times the weight of the polymer. Testing of the electrolyte in batteries has shown that alumina additions improve the cycling performance.

4. Conclusions

Solid electrolytes can provide advantages over liquid electrolytes in terms of safety, reliability and simplicity of design, but the ionic conductivities of solids are generally lower than those of liquids. However, both inorganic ceramics and organic polymers have been developed for use as the electrolytes in lithium-ion

batteries. The ranges of conductivities of some examples of these electrolytes are shown in Fig. 22. The highest conductivities are for LAGP and PVdF-HFP, but these have some disadvantages over other potential electrolytes, such as in mechanical strength or electrode compatibility. Selection of the most suitable electrolyte for a particular battery application depends on various factors, such as operating parameters (*e.g.* voltage range, temperature) and battery design (*e.g.* rigid or flexible).

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